Writhing Photons and Berry Phases in Polarized Multiple Scattering

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We study theoretically the polarization state of light in multiple scattering media in the limit of low contrast in the refractive index. Linearly polarized photons are randomly rotated due to the Berry phase associated with the scattering path. For circularly polarized light independent speckle patterns are found for the two helical states. The statistics of the geometric phase is related to the writhe distribution of semiflexible polymers such as DNA.

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Multiple light scattering techniques [1] allow the experimentalist to probe deep into opaque, strongly scattering samples and give information on both static and dynamic correlations. The preferred way of interpreting these experiments is via a field of *scalar* photons [2] which satisfy, in the simplest analytic treatments, a diffusion or transport equation. The phase of the wave associated with each path is proportional to the optical path length sampled by each photon. In this Letter, we show the importance of geometric phases in the evolution of photons in multiple scattering which have as their origin the vector nature of light. Rather surprisingly, while the basic propagation laws for light in inhomogeneous media go back over 60 yr to the work of Rytov [3,4], the full implication of his results has not yet been exploited in the interpretation of multiple scattering in condensed matter systems. In this Letter, we consider the consequences of Rytov's observations on the scattering of circularly and linearly polarized light.

We first summarize our main results before giving a more extended discussion: For *circularly polarized light,* the photon helicity is a conserved quantum number in systems with weak inhomogeneities [3] within the eikonal approximation. Each possible, multiple scattered, path through a sample is highly tortuous and thus has associated with it a writhe $\phi_i/2\pi$ [4,5]. A Berry phase, $e_i\phi_i$, then adds to the simple geometric optical path $\psi_i = q\ell_i$, calculated in a scalar theory, where q is a wave number, ℓ_i is the optical path length, and $e_i = \pm 1$ is the photon helicity. In a transmission geometry, this leads to two possible speckle patterns for a given arrangement of scatters as a function of the polarization state of the light. For *linearly polarized light* the situation is more subtle: For each individual path through the sample the geometric phase rotates the plane of polarization [6]. However, the final experimental measurement is a measure of intensity, summing over all paths through the sample. This sum leads to a final polarization state which is, in general, elliptical rather than linear. We shall thus characterize the evolution of a linearly polarized state as a probability distribution of polarization states on the Poincaré sphere.

Technically we treat the problem of summing over photon paths via a mapping onto a semiflexible polymer, which treats multiple weak scattering of photons as an *angular* diffusion process. The Berry phase is calculated from the writhe [7] of the photon path using methods introduced to study the statistical mechanics of DNA and other stiff molecules. Our considerations also link up with remarks of [8] which gave a local argument for the evolution of the polarization vector, in the context of backscattering, equivalent to that of Berry's.

Since multiple scattering techniques are very often used to study the properties of colloidal systems, we start by explaining how some of the above ideas are applied to such multicenter scattering systems, where the mapping onto a torsionally rigid semiflexible polymer is particularly direct and simple to understand. In the theory of scattering from colloidal samples, one defines two characteristic distances. The first, ℓ, is the distance between two collisions between a photon and a scattering center. The second, ℓ^*, measures the distance over which a photon must travel in order to forget its initial direction of propagation. In a simple analogy with stiff polymers, one can consider that the length ℓ corresponds to a monomer size while ℓ^* is equivalent to the persistence length of the polymer. In strongly scattering media, with high contrast between inclusions and background these two lengths are comparable. However, by using particles large compared with the wavelength of light and with low contrast between the dielectric properties of the two media, we can easily find samples for which ℓ^*/ℓ is of order 10. In recent experiments [9] in large droplet helium aerosols, it has proven possible to increase ℓ^*/ℓ to over 200. Motivated by this last experimental system, we present arguments as to the polarization statistics of light scattered in a regime of intermediate sample thickness, L, such that $\ell \ll L \leq \ell^*$ in transmission geometry. In such samples photons are scattered many times, but are still largely propagating in the forward direction.

We now derive the mapping which allows us to transpose results known from the statistical mechanics of stiff polymers, including now the torsion as well as bending degrees of freedom. Following [9], we shall model the medium as an ensemble of randomly oriented interfaces neglecting the spherical structure of the aerosol. This was shown to give a good qualitative description of the scattering

statistics. Consider the collision between a photon and a single interface. The photon can be either reflected, and thus deviated by a large angle, or refracted by a small angle which can be calculated using geometric optics. The probability of reflection, for a typical impact parameter, is comparable to $\mathcal{R} = (n-1)^2/(n+1)^2$, where n is the refractive index of the inclusions relative to that of the background medium. If we write $\epsilon = (n-1)$ then we see that the intensity of the direct beam decays over a length which is at most $\ell_{\rm ref} = \ell/\epsilon^2$ due to back scattering. A background of photons scattered through large angles is indeed observed experimentally, in addition to the main forward beam [9]. We shall ignore these photons in what follows.

Most of the beam is refracted at the interface. For typical impact parameters the angle of deviation is comparable to ϵ. Since successive deviations of a photon are independent, this implies that the direction of propagation of the photon diffuses as it propagates into the medium with angular diffusion coefficient $D \sim \epsilon^2/\ell$. Thus photons which are not directly reflected at a surface also turn over a length comparable to ℓ/ϵ^2 which is thus our estimate of ℓ^* in this weakly scattering limit. Note that in the case of spherical inclusions some extra care is needed. The intensity of scattering decays at large angles as $1/\theta^4$ [10]. The mean squared deviation has a logarithmic divergence and needs regularization by a cutoff or a treatment using generalized, Levy statistics since the central limit theorem does not apply.

Let us now consider the evolution of an incident linearly polarized beam. The reflectivity of an interface is a function of the plane of polarization with respect to the surface; the transmitted beam has a modified polarization state. In the hypothetical case of perfect transmission the plane of polarization of the refracted beam evolves by parallel transport [8,11]. Because of the reflections, the transmitted amplitudes of the two polarization components (defined relative to the local surface orientation) are comparable to $1 - O(\epsilon^2)$. This leads to a rotation of plane of polarization of the transmitted light by an angle $\pm O(\epsilon^2)$ compared to a parallel transported state. In the analogy with a stiff polymer this corresponds to excitation of a *torsional* mode. In stiff polymers, one can define two independent persistence lengths for bend and torsional degrees of freedom, l_p and l_t. These lengths are usually comparable. For the case of multiple light scattering, we see that $\ell_p \sim \ell^* \sim \ell/\epsilon^2$ whereas $\ell_t \sim \ell/\epsilon^4$. The "torsional" degree of freedom for photons is frozen out at low dielectric contrast so that as $\epsilon \to 0$ we find that $l_t \gg l_p$, and parallel transport of the transmitted component becomes exact.

The tangent to the propagation direction $\mathbf{t}(s)$ is a unit vector living on a unit sphere. We now use the Berry formula linking the area enclosed by the curve $\mathbf{t}(s)$ on this sphere with the geometric phase to calculate the probability distribution of phases associated with a samples in the limit $\ell \ll L \ll \ell^*$. When the sample is thin compared with ℓ^*, the photons do not diffuse very far from their original propagation direction. We can thus calculate the Berry phase by looking at the problem of diffusion on a local, planar approximation to the sphere. The probability distribution for the area of random loops on a plane is known [11,12], implying the probability distribution for the Berry phase: $\mathcal{P}(\phi) = 1/2DL\cosh 2(\phi/DL)$. From this expression we find the mean square phase as

$$\langle \phi^2 \rangle = \pi^2 L^2 D^2/12\,. \tag{1}$$

Because of the logarithmic divergence of the mean squared scattering angle for spherical droplets, we interpret this result as the typical writhing angle being linear in the sample thickness.

We proceed by studying the Jones vectors describing the electric field of a coherent light source. Vertically and horizontally polarized light is described by the vectors $j_v = (1,0)$ and $j_h = (0,1)$, respectively, whereas circular light corresponds to the vectors $j_\pm = 1/\sqrt{2}\,(1, \pm i)$. We are ultimately interested in the intensities of the various polarization states of the transmitted beam which are most easily visualized via the Poincaré sphere (Fig. 1). The three axes correspond to a series of measurements $i_1 = I_0 - I_{90}$, $i_2 = I_{45} - I_{-45}$, and $i_3 = I_+ - I_-$. Here, $I_{0,\pm 45,90}$ are the normalized intensities measured with a linear polarizer inclined at the subscripted angle, and $I_\pm$ is the intensity measured with circular analyzers.

As stated above for *circularly polarized light,* $e_i\phi_i$ can simply be added to the phase ψ_i so that the transmitted Jones vector is given by

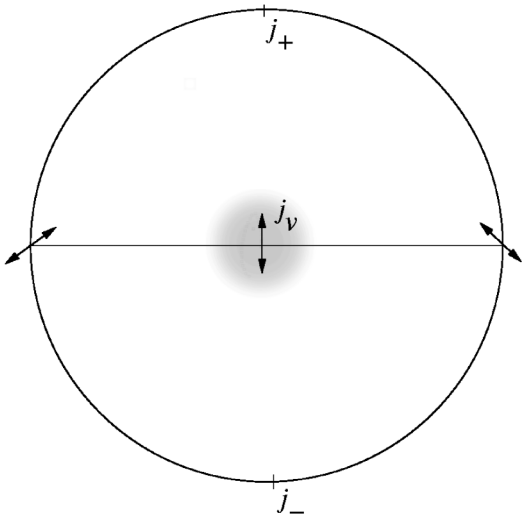

FIG. 1. The Poincaré sphere describing the polarization state of the light. Linearly polarized states correspond to points along the equator while the north and south poles, labeled j_+ and j_-, correspond to circularly polarized light. Other states are elliptically polarized. Multiple scattering of the initial state j_v describing linearly polarized light leads to a circularly symmetric distribution of polarization states centered on j_v in samples where $L/l* < 1$.

$$e^{i(\psi_i \pm \phi_i)} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix} \quad (2)$$

for the incident state $j_{\pm}$. We now sum over all possible paths to find the total scattered amplitude: The sum of the individual circularly polarized states also gives rise to circularly polarized light as the final state. When $L/l* \sim 2$ the geometric phase introduces a relative phase $2\phi_i$ between the right and the left states comparable to π. We thus understand that the speckle pattern for the two helical states is similar for very thin samples, whereas for thicker samples we find two independent intensity distributions for each circularly polarized state.

If we illuminate with *linearly polarized* light $j_v = (1, 0)$, each transmitted photon is described by its writhe ϕ_i and the total phase ψ_i so that the transmitted state is

$$e^{i\psi_i} \begin{pmatrix} \cos\phi_i \\ \sin\phi_i \end{pmatrix} \approx e^{i\psi_i} \begin{pmatrix} 1 \\ \phi_i \end{pmatrix}, \quad (3)$$

where we have specialized to samples with $L/\ell^* < 1$. ψ is calculated from the statistics of longitudinal fluctuation of a stiff polymer by writing the path length as $\Delta = \int ds\sqrt{1 + (d\mathbf{r}_\perp/ds)^2}$, with $\mathbf{r}_\perp$ the deviation of the path from a straight line. By expanding the square root and studying the correlation function $\langle \Delta^2 \rangle$ [13], one finds that the fluctuations of path length, ℓ_i, for a sample of thickness L are of order L^2/ℓ^*, when $L < \ell^*$. Thus ψ_i is very large, so that $\psi_i \mathrm{mod}(2\pi)$ is very nearly uniformly distributed.

If we sum the amplitude Eq. (3) over a large number of independent paths, corresponding to illuminating a large area of the sample, we find a vector of the form

$$j_f = A e^{i\psi_0} \begin{pmatrix} 1 \\ \phi_0 e^{i\psi_1} \end{pmatrix}. \quad (4)$$

A relative phase between the two components of the vector j_f develops because of the random sign of ϕ_i. For small, fixed ϕ_0, the vector j_f describes a circle of radius $2\phi_0$ on the Poincaré sphere as ψ_1 varies between 0 and 2π. We note, in passing, that the full joint distribution function of the writhe and path length, $\mathcal{P}(\phi, \psi)$, is closely linked with the force-torsion response curves measured in DNA micromanipulation experiments [14]. The joint distribution could be explored in optical experiments by performing time resolved studies of the polarization state with fast laser pulses.

Thus, A and $A\phi_0$ are random variables with $\phi_0 \sim \sqrt{\langle \phi^2 \rangle} \sim L/\ell^*$ [Eq. (1)]. ψ_1 is again uniformly distributed. For thin samples this random Jones vector is distributed in a circular disk, centered on the initial vector j_v (Fig. 1). We see that the radius of the disk is directly related to the typical writhe of paths through the sample. The combination of a Berry phase combined with the widely distributed optical path leads to a state of elliptical polarization. We expect a complete loss of memory of the initial polarization state in samples thicker than a few times ℓ^*.

In conclusion, we have reached an understanding as to the evolution of the polarization states in multiple scattering situations via a mapping onto a writhing polymer. Our results are valid for *coherent* light sources; the essential step was the combination of the *amplitudes* of the Jones vectors. In scattering with *incoherent* sources it is rather the Stokes parameters (i.e., the intensities, i_1, i_2, i_3, together with the total intensity i_0) which should be combined, presumably leading to multicomponent transfer theories such as those discussed in [15]. The contrast between multiple polarized scattering with coherent and incoherent sources deserves closer study.

We reiterate that there is an important *qualitative* difference between the evolution of circular and linear polarization states. For the former, the helicity can be preserved whereas the Berry phase associated with linearly polarized light always leads to a state of random polarization. We suggest that the natural setting for experimentally studying polarization effects in multiple scattering media is in critical, opalescent samples or in solutions of extremely large macromolecules, where the gradual density gradients allow an exact formulation of the polarization statistics without backscattering from interfaces.

In our rather crude description of the colloidal regime, we have neglected all diffraction, limiting our treatment to extremely large droplets. Many of the qualitative conclusions should hold even for more moderate particle sizes in Rayleigh-Gans scattering where diffraction is important. Detailed simulations [16,17] have been performed in this regime to study the decay of polarization in colloidal systems in transmission geometry with parameters corresponding to polystyrene beads in water. It was indeed observed numerically that the evolution of the polarization state proceeds by the formation of a circular distribution on the Poincaré sphere. The authors defined new lengths $\ell^{**}_{\mathrm{circ}}$ and $\ell^{**}_{\mathrm{plane}}$, over which both circular and linear polarization decay while noting that $\ell^{**}_{\mathrm{circ}} > \ell^{**}_{\mathrm{plane}}$. It would be particularly interesting to perform simulations in the limit of very low optical contrast in order to bring out some of the scaling regimes which may exist in this limit.

In lower symmetry samples, rather different results are to be expected. Light propagating in a *uniform* birefringent material is described by two refractive indexes describing a slow and a fast wave. In such a material circularly polarized light can no longer propagate without modification. In the presence of both multiple scattering and birefringence, we expect that there is *no* stable state of polarization. It is to be noted that many tissues, such as muscle, do have significant birefringence. Empirically it has been found that deep optical imaging in colloids is often best achieved with circularly polarized light which we understand eliminates polarization modifications due to writhe; in tissues linearly polarized light is preferred [18].

Finally, we have not considered the nature of the non-Gaussian statistics of the polarization states due to the caustics and imaging presumably present in weak

scattering limits discussed in this Letter. A final question that we leave open is the nature of two time correlation functions of the polarization state: As scattering centers move both the writhe of the photon paths and the phase associated with each path vary. Does this variation contain any interesting information on the dynamics of colloidal systems?

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